O Docket No. <u>1776-4</u>066

## Appendix 2

## **New Claims**

--87. A hydroxyalkyl(meth)acrylate composition comprising:

0.3 to less than 1.0 mole of polymerized lactone monomer being polymerized by ring-opening with respect to 1 mole of hydroxyalkyl(meth)acrylate, wherein the content of the lactone monomer in the composition is 0-10% by weight, and a proportion of monomers having two or more continuous chains (n ≥ 2) of lactones less than 37.4% (area by GPC),

the composition being represented by formula (1) described below,

where

R, R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> are independently a hydrogen or a methyl group,

"j" is an integer of 2-6,

xn pieces of R<sup>4</sup> and R<sup>5</sup> are independently a hydrogen or an alkyl group having a carbon number of 1-12,

"x" is 4-7,

"n" is an integer greater than or equal to zero, and

an average value of "n" in the composition is not less than 0.3 to less than 1.0.

88. A method for the preparation of a hydroxylalkyl(meth)acrylate composition comprising a small amount of lactones, said method comprising:

reacting, per mole of hydroxyalkyl(meth)acrylate, 0.3 to less than 1.0 mol of a

lactone by ring-opening polymerization according to the reaction represented below:

(where R, R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> are independently a hydrogen or a methyl group, "j" is an integer of 2-6, xn pieces of R<sup>4</sup> and R<sup>5</sup> are independently a hydrogen or an alkyl group having a carbon number of 1-12, "x" is 4-7, "n" is an integer equal to 0 or greater, and an average value of "n" in a composition is not less than 0.3 to less than 1.0), so as to prepare a polylactone-modified hydroxyalkyl(meth)acrylate composition wherein a proportion of monomers having not less than 2 continuous chains (n  $\geq$  2) of lactones is less than 37.4% (GPC area %) in the composition and the content of the lactone monomer remaining in the composition is 0-10% by weight.--

## **Amended Claims**

2. A hydroxyalkyl(meth)acrylate composition [modified by] comprising a small amount of lactones as claimed in claim [1] 87, wherein a hydroxyalkyl(meth)acrylate which is employed as a raw material is a hydroxyethyl(meth)acrylate.

- 3. A hydroxyalkyl(meth)acrylate composition [modified by] comprising a small amount of lactones as claimed in claim [1] <u>87</u>, wherein a lactone monomer which is employed as a raw material is ε-caprolactone and/or valerolactone.
- 4. A hydroxyalkyl(meth)acrylate composition [modified by] <u>comprising</u> a small amount of lactones as claimed in claim [1] <u>87</u>, wherein the content of a lactone monomer [remained] remaining in the composition is 0-10% by weight.
- 5. A hydroxyalkyl(meth)acrylate composition [modified by] comprising a small amount of lactones as claimed in claim [1] 87, wherein the content of a hydroxyalkyl(meth)acrylate [remained] remaining in the composition is not less than 20% by weight and not more than 50% by weight.
- 6. A hydroxylalkyl(meth)acrylate composition [modified by] <u>comprising</u> a small amount of lactones as claimed in claim <u>87</u> [1], wherein the contents of a di(meth)acrylate which is a by-product in said composition is not more than 2% by weight.
- 7. A hydroxyalkyl(meth)acrylate composition [modified by] comprising a small amount of lactones as claimed in claim [1] <u>87</u>, wherein the [content of] by-products produced by [side reactions such as a] Michael addition, [an] acrylic polymerization, [a] transesterification, and other side reactions is not more than 10% by weight in said composition.

- 12. A method for the preparation of a hydroxyalkyl(meth)acrylate composition [modified by] comprising a small amount of lactones as claimed in claim [11] <u>88</u>, wherein said hydroxyalkyl(meth)acrylate is hydroxymethylacrylate or hydroxyethylmethacrylate.
- 21. A curable resin composition comprising 0.5 80 parts by weight of an acrylic polyol resin (A) obtained using
  - [a] the hydroxyalkyl(meth)acrylate composition [(a) modified by a small amount of lactones represented by the above-described formula (1)] as claimed in claim [1]  $\underline{87}$ , in which a proportion of monomers having not less than 2 continuous chains ( $n\geq 2$ ) of lactones is less than 50% (GPC area %), as polymerizing components, and
  - 0.5 parts by weight of a melamine resin (B), the total of (A) and (B) not exceeding 100 parts by weight.
- 22. A curable resin composition as claimed in claim 21, wherein said hydroxyalkyl(meth)acrylate composition [(A) modified by a small amount of lactones] is obtained using a hydroxyethyl(meth)acrylate.
- 26. A melamine-curable, [type] water-based coating composition comprising:
  - 5-30 parts by weight of an acrylic polyol resin (A) obtained using a hydroxyalkyl(meth)acrylate composition [(a) modified by a small amount of lactones represented by the above-described general formula (1)] as claimed in claim <u>87</u>, wherein a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is less than 50% (GPC area

%)[,] as polymerizing components, and 10-60 parts by weight of an amino-plasto resin (IV-B).

- 27. A melamine-curable, [type] water-based coating composition as claimed in claim 26, [characterized by composing of] <u>further comprising:</u>
  - (i) 3-40% by weight of the hydroxyalkyl(meth)acrylate composition[ (a) modified by a small amount of lactones],
  - (ii) 1-20% by weight of  $\alpha,\beta$ -unsaturated carboxylic acid,
  - (iii) 1-25% by weight of an N-alkoxymethyl(meth)acrylate having a carbon number of 1-6 in an alkyl group, and
  - (iv) an aromatic vinyl monomer and an alkyl(meth)acrylate which are contained in an amount that <u>is equal to</u> 100% by weight minus total weight % of the above components (i), (ii), and (iii).
- 31. A curable resin composition comprising:
  - 50-90 parts by weight of an acrylic polyol resin (V-A) obtained using a hydroxyalkyl(meth)acrylate composition (a) [modified by] comprising a small amount of lactones, represented by the above-described general formula (1) as claimed in claim [1] 87, in which a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is less than 50% (GPC area %), and

50-10 parts by weight of a polyisocyanate compound (V-B), wherein the total of the (V-A) and (V-B) does not exceed[ing] 100 parts by

weight.

- A) is composed of 5-65% by weight of the hydroxyalkyl(meth)acrylate composition (a).

  [modified by a small amount of lactones,] 0-30% by weight of a vinyl monomer having a hydroxyl group, and other vinyl-based monomers (residual weight).
- 33. A curable resin composition comprising 0.5 80 parts by weight of an acrylic polyol resin (VI-A) having a carboxylic group and a functional group obtained by [allowing to] reacting a hydroxyalkyl(meth)acrylate composition [(a) modified by a small amount of lactones,] as claimed in claim 87 with

a vinyl monomer having carboxylic group and other vinyl monomers, and 0.5 – 50 parts by weight of a polyisocynate compound (VI-B),

[represented by the general formula (1) claimed in claim 1,] wherein a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is less than 50% (GPC area %), [with a vinyl monomer having carboxylic group and other vinyl monomers, and 0.5 – 50 parts by weight of a polyisocynate compound (VI-B)], and the total of the (VI-A) and (VI-B) does not exceed[ing] 100 parts by weight.

34. A curable resin composition as claimed in claim 33, wherein said acrylic polyol resin (VI-A) is a vinyl-based copolymer having a carboxylic group and a functional group, said copolymer being obtained by [allowing to react] reacting a reaction product of a hydroxyl group-[contained]containing resin and a (meth)acrylic anhydride with a vinyl-based

monomer having carboxylic group and other vinyl based monomers, said hydroxyl group-[contained]containing resin being obtained by polymerizing [a] the hydroxyalkyl(meth)acrylate composition (a) [modified by a small amount of lactones].

- 35. A curable resin composition as claimed in claim [33] 34, wherein said acrylic polyol resin (VI-A) [is a resin obtained using] comprises said hydroxyl group[contained]containing resin [as claimed in claim 34] and at least one resin selected from a group consisting of a urethane resin having hydroxyl groups, an epoxy resin having hydroxyl groups, a cellulose derivative having hydroxyl groups, and a polyester resin having hydroxyl groups.
- 36. A curable resin composition as claimed in claim 33 wherein said acrylic polyol resin (VI-A) is a resin [using] comprising said hydroxyl group-[contained]containing resin [claimed in claim 34] resin obtained by polymerizing the hydroxyalkyl(meth)acrylate composition and a urethane resin having hydroxyl groups.
- 39. A coating [characterized by containing] <u>comprising</u>:
  - (i) crosslinked particles obtained by dispersing a mixture of an acrylic polyol resin (VI-A) with a polyisocyanate compound (VI-B) into a water-based medium and by crosslinking thereof, or
  - (ii) composite[-type] crosslinked particles composed of a urethane-urea/ethylene-based resin obtained through polymerizing polymerizable ethylenic unsaturated compounds containing a hydroxyalkyl(meth)acrylate composition [(a) modified by]

comprising a small amount of lactones in water in which crosslinked urethane-urea particles are dispersed, as resin components for forming a thin layer.

- 40. A coating as claimed in claim 39, [characterized by containing] <u>further comprising</u> not less than 50% by weight of crosslinked particles having <u>a</u> particle diameter of not more than 1 μm and, moreover, an average molecular weight between crosslinking points within a range of 300-2,000, as resin components for forming a thin layer.
- 46. A thermosetting resin composition which comprises 2-50 parts of an acrylic polyol resin (VII-A) containing a hydroxyalkyl(meth)acrylate composition [(a) modified by a small amount of lactones represented by the general formula (1)] as claimed in claim [1] <u>87</u>, wherein a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is less than 50% (GPC area %), and 30-80 parts of an acrylic copolymer (VII-B) having an alkoxylsilyl group, total of [the] (VII-A) and (VII-B) being 100 parts by weight.
- 47. A thermosetting resin composition as claimed in claim 46, wherein said acrylic polyol resin (VII-A) has at least one kind of group selected from the group consisting of an acid anhydride group, an epoxy group, an amino group, and a carboxylic group.
- A method for the preparation of a carboxylic group-[contained]containing acrylate composition (a') [modified by a small amount of lactones] represented by a general formula (VIII-3) described below [characterized by allowing to react], said method

Docket No. 1776-4066

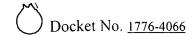
comprising:

reacting the [a] hydroxyalkyl(meth)acrylate composition [(a) modified by a small amount of lactones represented by the above-described formula (1)] <u>as</u> claimed in claim [1] <u>87</u>, in which a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is less than 50% (GPC area %), with a carboxylic acid or anhydride thereof (VIII-b) represented by a general formula (VIII-2) described below,

$$R^9 - [COOH]_{m+1}$$
 (VIII-2)

(in the formula, R, R<sup>1</sup>, R<sup>2</sup>, and R<sup>3</sup> are independently a hydrogen or a methyl group, "j" is an integer of 2-6, xn pieces of R<sup>4</sup> and R<sup>5</sup> are independently a hydrogen or an alkyl group having a carbon number of 1-12, "x" is 4-7, "n" [is 0 or an integer of not less than 1] is an integer greater than or equal to zero, an average value of "n" in said composition is not less than 0.3 to less than 1.0, R<sup>9</sup> is a residual group of a carboxylic acid, and "m" is an integer of 1-3).

56. A method for the preparation of a carboxylic group-[contained]containing acrylate composition (a') [modified by a small amount of lactones] as claimed in claim 55,



[characterized in that a] wherein said reaction of said hydroxyalkyl(meth)acrylate composition (a) [modified by] comprising a small amount of lactones with said carboxylic acid or anhydride thereof (VIII-b) is conducted at a temperature range of 40 – 160 °C.

59. A curable resin composition which comprises 10-70 parts of an acrylic polycarboxylic acid resin (A') [containing] comprising:

[said] the carboxylic group-[contained]containing acrylate composition (a'), [modified by] comprising a small amount of lactones, represented by the general formula (VIII-3) as claimed in claim 55, in which a proportion of monomers having not less than 2 continuous chains ( $n\geq 2$ ) of lactones is less than 50% (GPC area %), as a polymerizing component, and

10-80 parts of a polyepoxide (IX-B).

60. A curable resin composition as claimed in claim 59, [characterized in that] wherein said carboxylic group-[contained]containing hydroxy(meth)acrylate composition (a') [modified by a small amount of lactones] is obtained by allowing to react said hydroxyalkyl(meth)acrylate composition, [modified by a small amount of lactones, represented by the general formula (1) as claimed in claim [1] 87,] in which a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is less than 50% (GPC area %), with said carboxylic acid or anhydride thereof represented by the following general formula [(VIII-2) as claimed in claim 55.]:

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Serial No. 09/890,321

Docket No. 1776-4066

R9 -[COOH]<sub>m+1</sub>

wherein R<sup>9</sup> is a residual group of a carboxylic acid, and "m" is an integer of 1-3.

- 61. A curable resin composition as claimed in claim 60, [characterized in that] wherein said carboxylic group-[contained]containing hydroxy(meth)acrylate composition (a') [modified by] comprising a small amount of lactones is obtained by allowing to react 0.9 1.1 mol of said carboxylic acid or anhydride thereof with respect to 1 mole of said hydroxy(meth)acrylate composition (a) [modified by] comprising a small amount of lactones.
- 71. A polyester unsaturated monomer composition [modified by] <u>comprising</u> a small amount of lactones, in which a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is less than 50% (GPC area %), [which is] <u>said composition</u> obtained by a ring-opening polymerization of <u>less than 1 mole of</u> a lactone monomer with respect to [a] <u>1 mole of</u> polymerizable unsaturated monomer containing <u>a carboxylic group</u>.
- 72. A polyester unsaturated monomer composition [modified by] <u>comprising</u> a small amount of lactones as claimed in claim 71, wherein said polymerizable unsaturated monomer containing carboxylic group is at least one kind selected from the group consisting of a (meth)acrylic acid, itaconic acid, β-(meth)acryloyloxyethyl succinic acid, β-(meth) acryloyloxyethyl maleic acid, β-(meth)acryloyloxyethyl phthalic acid, maleic acid, a monoalkyl maleate (a carbon number in an alkyl group is 1-12), tetrahydrophthalic acid, and an anhydride thereof.

73. A polyester unsaturated monomer composition [modified by a small amount of lactones] as claimed in claim 72, wherein said polymerizable unsaturated monomer containing carboxylic group is a (meth)acrylic acid, and which is obtained by a reaction represented by (X –1) described below,

$$\begin{array}{ccc}
R & R^1 & R^4 \\
C = CCOO((C)xCOO)n-H & (X-1)\\
R^5
\end{array}$$

(in the formula, R and R<sup>1</sup> are independently a hydrogen or a methyl group, xn pieces of R<sup>4</sup> and R<sup>5</sup> are independently a hydrogen or an alkyl group having a carbon number of 1-12, "x" is 4-7, "n" and "n" in said composition are [0 or an integer or not less than 1] an integer greater than or equal to zero, and an average value of "n" in said composition is not less than 0.3 to less than 1.0).

82. A method for the preparation of a polyester unsaturated monomer composition, [modified by] comprising a small amount of lactones, wherein 0.3-less than 1.0 mole of a lactone monomer is polymerized by ring-opening with respect to 1 mole of a radically polymerizable unsaturated monomer[s] containing carboxylic group, whereby, a proportion of monomers having not less than 2 continuous chains (n≥2) of lactones is adjusted to less than 50% (GPC area %).

- 83. A method for the preparation of a polyester unsaturated monomer composition [modified by] comprising a small amount of lactone as claimed in claim 82, wherein an acidic catalyst is a Lewis acid or a [Br nsted] Brønsted acid.
- 85. A method for the preparation of a polyester unsaturated monomer composition [modified by a small amount of lactone, characterized in that] <u>comprising</u>:

polymerizing, by ring opening, 0.3 – less than 1.0 mole of a lactone monomer [ is polymerized by ring-opening] with respect to 1 mole of a radically polymerizable unsaturated monomer containing carboxylic group by using stannous halide, monobutylin tris-2-ethylhexanate, stannous octoate, dibutylin dilaurate, or a mixture thereof as a catalyst, and [followed by]

separating the unreacted radically polymerizable unsaturated monomer containing carboxylic group.

86. A method of the preparation of a polyester unsaturated monomer composition [modified by a small amount of lactone] as claimed in claim 85, wherein the catalyst to be employed in said [ring-opening] polymerization is less than 1000 ppm by weight based on total amount to be fed.